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Fracton contribution to the temperature dependence of the homogeneous linewidth of the $^5D_0$-$^7F_0$ transition in Eu^{3+}-doped glasses

George S. Dixon, Richard C. Powell, and Xu Gang
Department of Physics, Oklahoma State University, Stillwater, Oklahoma 74078

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Laser-induced fluorescence line-narrowing techniques were used to measure the homogeneous linewidth of the $^5D_0$-$^7F_0$ transition in Eu^{3+}-doped glass hosts after resonant excitation. The homogeneous linewidths were measured as a function of temperature between 10 and 300 K and as a function of excitation wavelength across the inhomogeneous bands in two types of silicate and one germanate glass. The residual inhomogeneous linewidth at 10 K increases linearly with energy across the band and the homogeneous width at both high- and low-energy sides of the inhomogeneous band exhibits a T^2 temperature dependence over the entire range investigated. A model based on the fracton concept for the anomalous vibrational degrees of freedom of the glass hosts is developed to explain the observed behavior of the temperature-dependent line broadening.

I. INTRODUCTION

Physical properties such as thermal conductivity, heat capacity, and ultrasonic attenuation are known to be quite different at low temperatures in glasses as compared to crystals.[1] This has been attributed to additional degrees of freedom in amorphous solids that are not present in crystals. These are described by a phenomenological model consisting of two possible local atomic or bonding equilibrium arrangements separated by a potential barrier.[2,3] The local system can vibrate within one potential well or tunnel between the two wells. These localized "two-level systems" (TLS) provide the additional density of states necessary to interpret many of the observed experimental results. However, there are still many unanswered questions concerning the fundamental nature of these TLS and the role they play in determining other properties of amorphous solids. Thus, this is still a very active area of research. Two interesting questions concern the importance of the TLS modes at higher temperatures and the possible variation of TLS properties for different types of glasses.

One method of probing the properties of motional degrees of freedom of solids has been to study the temperature dependence of the homogeneous linewidth of an optical transition of an impurity ion in the solid. This has become an especially powerful tool with the use of high-resolution lasers to selectively excite subsets of ions within an inhomogeneously broadened spectral profile so homogeneous linewidths can be measured even in the presence of significant static strain broadening. Hole burning in absorption and fluorescence line narrowing (FLN) in emission make use of this technique. FLN experiments on Eu^{3+} ions in a silicate glass host have shown a T^2 dependence for the homogeneous linewidth at low temperatures.[4] This has been interpreted in terms of contributions to the line broadening due to the interaction between the impurity ion and the local TLS.[5-7] Further experimental data are required to increase our understanding of this type of line-broadening mechanism.

We report here the results of FLN measurements on Eu^{3+} in three different types of glass hosts at temperatures ranging from 10 to 300 K. The residual inhomogeneous linewidth at 10 K is found to increase linearly with energy across the inhomogeneously broadened band. The homogeneous linewidth at both the high- and low-energy sides of the inhomogeneously broadened band is found to obey a T^2 dependence over the entire temperature range investigated for all three glasses.

It was first suggested by Stapleton and co-workers[8] that fractal structures might have an anomalous phonon density of states that would affect electron-phonon relaxation processes. Using the fracton concept of Alexander and Orbach,[9] we consider the coupling of the impurity ion to the vibrational degrees of freedom of the amorphous host. Both the temperature dependence and the magnitude of the observed linewidths are consistent with Raman broadening by the localized vibrations (fractons).

II. EXPERIMENTAL

The laser-narrowed fluorescence linewidth was measured with the apparatus shown schematically in Fig. 1. The 10 ns excitation pulse from the nitrogen laser-pumped tunable dye laser passed through an extra-cavity étalon reducing its linewidth to 0.10 cm^{-1}. Coumarin dye was used to produce resonant excitation of the Eu^{3+} $^7F_0$-$^5D_0$ transition of interest. The samples were mounted on the cold finger of a closed-cycle helium cryostat permitting measurements between 10 and 300 K. The fluorescence was focused on the entrance slit of a Spex 1-m spectrometer set for a resolution of 0.08 Å. A shutter placed between the sample and the spectrometer was opened 0.6 ms after the excitation pulse to eliminate the possibility of contamination of the signal by scattered laser light after resonant excitation. The window of the boxcar integrator was set to look at the spectrum 1.0 ms after the excitation pulse. It is shown elsewhere that energy transfer does not broaden the fluorescence on this time scale.[10]

Three europium-doped glass samples were investigated. These had the following composition in mole per cent:

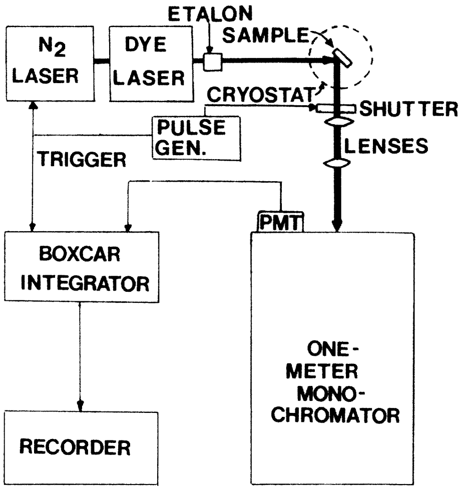

FIG. 1. Bloch diagram of experimental setup.

lithium silicate glass (LS), 57.0 SiO_2, 27.5 Li_2O, 10.0 CaO, 2.5 Al_2O_3, 3.0 Eu_2O_3; sodium silicate glass (NS), 72.0 SiO_2, 15.0 Na_2O, 5.0 BaO, 5.0 ZrO, 3.0 Eu_2O_3; and potassium germanate glass (KG), 63.3 GeO_2, 17.0 K_2O, 17.0 BaO, 0.7 Eu_2O_3. The spectral properties of these samples have been reported previously.[10]

III. HOMOGENEOUS AND INHOMOGENEOUS CONTRIBUTIONS TO THE LASER-NARROWED FLUORESCENCE LINEWIDTH

For resonant excitation of a transition, such as that studied here, the transition linewidth is given using the approximation of Kushida and Takushi,[11]

$$2\Delta\nu=\Delta\nu_{\text{obs}}-\Delta\nu_{\text{res}}\ , \qquad (1)$$

where $\Delta\nu$ is the linewidth of the transition, $\Delta\nu_{\text{obs}}$ is the observed fluorescence linewidth, and $\Delta\nu_{\text{res}}$ is the instrumental resolution. For our setup the monochromator resolution is less than 0.05 cm^{-1}, so $\Delta\nu_{\text{res}}$ is determined by the étalon-narrowed laser pulse width, which is 0.1 cm^{-1}. Figure 2 displays the temperature dependence of the narrowed fluorescence linewidth for excitation on the high- and low-energy sides of the inhomogeneously broadened band for the lithium silicate glass, the sodium silicate glass, and the potassium germanate glass. In each case the linewidth has an approximately quadratic temperature dependence at high temperatures, but is seen to approach a temperature-independent limit at low temperatures. This is different from previously reported results in glasses[4] where the quadratic temperature dependence was seen to extend to 10 K. Note that the limiting values of the linewidths in Fig. 2 are about an order of magnitude larger than the instrumental resolution. However, because we have used an extra-cavity étalon rather than an intracavity étalon as employed by Yen and co-workers,[4,12] the linewidth of our excitation pulse is an order of magnitude larger. This suggests that the temperature-independent contribution to the linewidth is residual inhomogeneous broadening due to imperfect site selection. Because the inhomogeneous contribution has been reduced to about 1 cm^{-1} from the several hundred cm^{-1} width before site selective excitation,[10] it can be deconvoluted from the homogeneous linewidth using the same techniques employed for crystalline hosts. The results of this procedure are shown in Fig. 3. They are consistent with a quadratic temperature dependence for the homogeneous linewidth from 10 K to the highest temperatures measured for all of the samples.

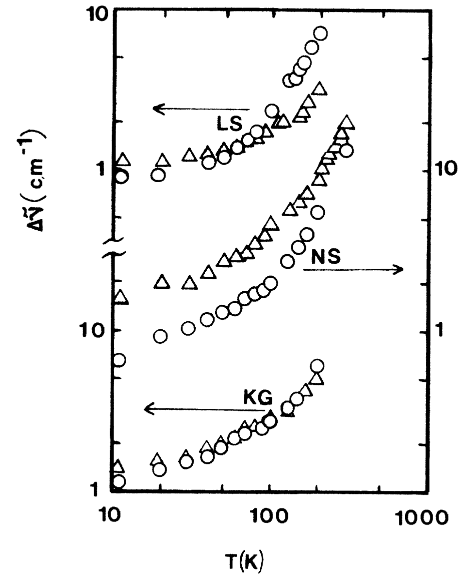

FIG. 2. Temperature dependence of the laser-narrowed linewidths of the ${}^5D_0$-${}^7F_0$ transition of Eu^{3+} in LS, NS, and KG glasses excited on the high-energy ($\lambda=577.5$ nm, triangles) and low-energy ($\lambda=580.0$ nm, circles) sides of the inhomogeneously broadened band.

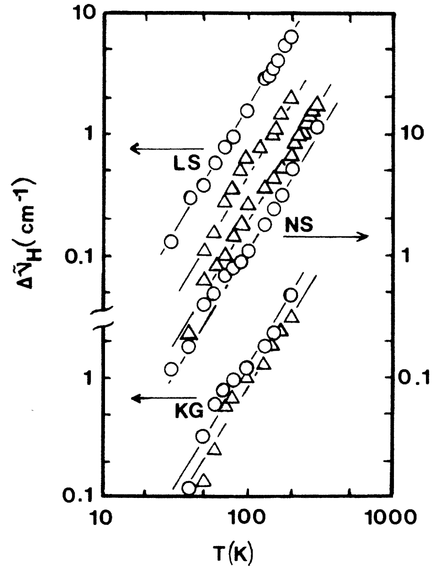

FIG. 3. Temperature dependence of the homogeneous linewidths of the ${}^5D_0$-${}^7F_0$ transition of Eu^{3+} in LS, NS, and KG glasses excited on the high-energy ($\lambda=577.5$ nm, triangles) and low-energy ($\lambda=580.0$ nm, circles) sides of the inhomogeneously broadened band.

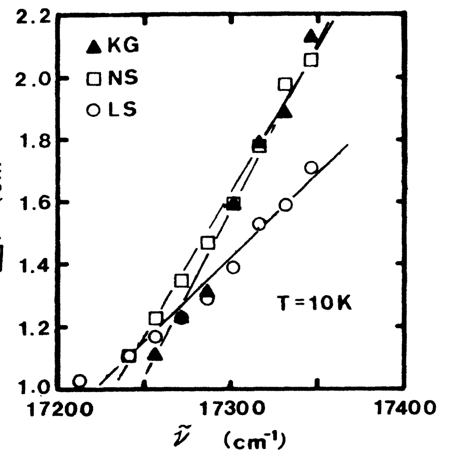

FIG. 4. Residual inhomogeneous broadening of the $^5D_0$-$^7F_0$ transition of Eu^{3+} in LS, NS, and KG glass at 10 K as a function of excitation wave number across the inhomogeneously broadened band.

The dependence of the linewidth on the excitation frequency was also measured at 10 K. These results are shown in Fig. 4. It can be seen that the linewidth increases monotonically in a roughly linear fashion as the excitation frequency is scanned from the low- to the high-frequency side of the inhomogeneously broadened band. This is the behavior to be expected of the residual inhomogeneous linewidth for the following reason. The inhomogeneous broadening arises from variations in the crystal field from site to site in the glass. The residual broadening is a measure of the number of different local environments that have crystal fields similar enough that the energy difference in their electronic transitions is within the excitation linewidth. One naively expects this density-of-site distribution to be greater on the high-energy side of the inhomogeneously broadened band where the crystal field splitting is strongest due to the larger number of ligands involved. It is noteworthy that the *homogeneous* linewidth does not behave in this way in one of the glasses studied. Figures 2 and 3 show that for the LS glass the measured linewidth is smaller on the high-frequency side of the band than on the low at temperatures above 60 K. The homogeneous linewidth which we extract from the measurements is smaller on the high-frequency side at all temperatures covered in these experiments. This behavior of the homogeneous linewidth is opposite to that of the other two glasses studied and to the observations of Morgan *et al.*[7] on the linewidth of a variety of glasses at room temperature.

IV. FRACTON MODEL FOR THE HOMOGENEOUS LINEWIDTH

In crystals the electron-phonon interactions control the temperature-dependent linewidth, with direct processes producing an exponential dependence and Raman processes leading to a T^7 dependence for $T \ll \Theta_D$ and a T^2 dependence for $T \gg \Theta_D$.[12,13] This is quite different from glasses which exhibit a universal quadratic temperature dependence for the homogeneous linewidth. Lyo and Orbach[5] have shown that the TLS that have been invoked to explain the anomalous thermal properties of glasses below 1 K can lead to a quadratic dependence of the homogeneous linewidth. However, this result holds as a low-temperature approximation only and requires a uniform density of states for the TLS. At high temperatures this model produces a linear temperature dependence. It is difficult to accept that the low-temperature limit would still be valid at 300 K. Although the TLS is a very successful model in accounting for thermal properties at very low temperatures and acoustic properties at low frequencies, the nature of these TLS has not been determined. While it is generally accepted that they involve tunneling between neighboring sites of local equilibria in the glass structure, it is not known whether the entities involved in the tunneling are individual ions or much larger clusters. If the latter were the case, their effect on the local environment of the optically active ions might be negligible.

In this section we develop a simple model of the homogeneous linewidth based on the "fracton" concept of Alexander and Orbach.[9] This differs from the TLS model in that the impurity ion interacts with the vibrational excitations of the host rather than the TLS modes. The quadratic temperature dependence is shown to be a natural result of Raman processes involving these non-Debye-type atomic vibrations and the magnitudes of the electron-phonon (or electron-fracton) coupling constants needed to obtain the observed linewidths are found to be consistent with those found in crystals.

The Raman contribution to the homogeneous linewidth is given by

$$\Delta\nu_R = (4\pi^2/\hbar)\,|\langle f\,|\,V_R\,|\,i\rangle|^2 \times \int_0^{\omega_{\max}} |\langle f\,|\,\epsilon^2\,|\,i\rangle|^2 g(\omega)^2 d\omega \;, \tag{2}$$

where $\langle f\,|\,\epsilon^2\,|\,i\rangle$ is a matrix element of the average local strain, $g(\omega)$ is the vibrational density of states, and $\langle f\,|\,V_R\,|\,i\rangle$ is the electrostatic matrix element for the Raman process. With the strains and the density of states treated in the Debye approximation, this leads to the conventional McCumber and Sturge result for crystals.[13] The expression shown in Eq. (2) is based on a perturbation theory approach to the problem. Recently a nonperturbative theory has been developed by Hsu and Skinner[14] which gives the same result as the perturbative approach in the weak coupling limit. At low temperatures both theories have the same form of the expression describing the linewidth with a different meaning for the coupling coefficients. Since no exact values are known for parameters such as the Debye temperature and electron-phonon coupling parameters for the samples used in this work, we use the form of the expression consistent with weak coupling and treat the Debye temperature and coupling parameters as adjustable parameters.

In glasses the Debye approximation is not a good representation of the vibrational states except at very long wavelengths. Orbach and co-workers[9,15] have shown that for shorter wavelengths, still much larger than atomic dimensions, plane waves are no longer a good representation of the normal modes; instead, they are localized in the disordered structures of the glass network. By modeling the glass structure as a self-similar fractal, Alexander and Orbach[9] have been able to obtain the vibrational density of states and dispersion without recourse to the usual reciprocal space transformations. In a series of papers[15] Orbach and co-workers have shown that this model can explain the thermal properties of amorphous solids above 1 K. Below this temperature it is still necessary to invoke the elusive TLS. In their model the vibrations at long wavelengths are treated in the Debye approximation, but at some critical length scale L, the disorder in the network causes the vibrations to become localized. The resulting vibrational density of states is

$$\begin{aligned} g_P(\omega) &= d(d/L^d)\omega^{d-1}/\omega_D^d, \quad l > L \\ g_F(\omega) &= \tilde{d}(d/L^d)(L/a)^{\bar{d}}\omega^{\tilde{d}-1}/\omega_F^{\tilde{d}}, \quad l < L , \end{aligned} \tag{3}$$

where ω_D is the effective "Debye frequency" for the phonons at $l=L$, ω_F is the upper limit for the fracton frequency (chosen to normalize the mode counting), a is the interatomic spacing, and d, $\bar{d}$, and $\tilde{d}$ are, respectively, the Euclidean, Hausdorff, and fracton dimensionalities. The fracton frequency at $l=L$ is ω_L. The characteristic length l of the vibration is its wavelength in the plane-wave regime and its localization length in the fracton regime. For the fractons the dispersion is no longer Debye-type. Assuming only a single kind of self-similarity,

$$\omega \approx l^{-\bar{d}/\tilde{d}} . \tag{4}$$

This might not be the case if there were several different characteristic lengths in the glass characterizing, for example, density and bond strength, or the same property in different parts of the glass.

One expects rather generally that the average local strain associated with a normal mode of amplitude Q should scale as Q/l. With this, the dispersion relationship, and density of states given above, the Raman contribution to the linewidth becomes

$$\begin{aligned} \Delta\nu_R = {} & \alpha(T/\Theta_D)^7 \int_0^{\Theta_D/T} x^6 e^x (e^x-1)^{-2} dx \\ & + A(T/\Theta_L)^{p+1} \int_{\Theta_L/T}^{\Theta_F/T} x^p e^x (e^x-1)^{-2} dx , \end{aligned} \tag{5}$$

where each Θ_i is $\hbar/k$ times the corresponding ω_i and the exponent $p=4(\tilde{d}/\bar{d})+2\tilde{d}-4$. The α and A parameters contain the same $|\langle f \mid V_R \mid i \rangle|^2$ electron-phonon coupling parameter as in the conventional treatment for crystals. For the glasses studies here $d=3$. The Hausdorff dimensionality $\bar{d}$ must lie between 3 and 2.5, which is the value found for a critically percolating structure. The fracton dimensionality $\tilde{d}$ was first conjectured by Alexander and Orbach[9] to be universally $\frac{4}{3}$. Recent calculations in the effective-medium approximation by Derrida *et al.*[15] give $\tilde{d} \approx 1$; however, this technique frequently does not give trustworthy estimates of exponents. Θ_D will now be much smaller than crystalline Debye temperatures. It is well known that for $T > \Theta_D/2$ the Debye-phonon term in Eq. (5) becomes quadratic in T. In Fig. 4 we show the results of a numerical evaluation of the fracton contribution to the linewidth. This can be seen to approach T^2 for any of the dimensionalities in the expected range, provided $T > 2\Theta_L$.

The structure of the host glass enters this model through the Hausdorff dimensionality $\bar{d}$. If $\bar{d} \approx d$, the structure is compact and has essentially the same atomic connectivity as would a crystal. An open structure for the glass will have a smaller value for $\bar{d}$, limited of course by $\bar{d}=2.5$ for a critically percolating cluster. The results in Fig. 5 are not especially sensitive to $\bar{d}$. Thus, assuming a nearly universal value for $\tilde{d}$ as conjectured by Alexander and Orbach,[9] the linewidth should not depend strongly on the glass structure.

In order to fit the specific-heat data above 1 K, Tua *et al.*[15] find $\Theta_L = 14$ K, $\Theta_D = 29$ K, and $\tilde{d} = 1.3$. Assuming similar values for the glasses studied here, an essentially quadratic temperature dependence for the homogeneous linewidth is predicted throughout the range covered by these experiments. The fracton contribution will dominate the Debye-phonon contribution at all temperatures in this range.

A first-principles calculation of the electron-phonon coupling $|\langle f \mid V_R \mid i \rangle|^2$ is not presently possible even for crystals. One can, however, calculate the A parameter required to produce the experimentally observed linewidth and use this to estimate the α parameter that would be observed in a crystal with a Debye-phonon spectrum and the same average sound velocity and density as the glass. This equivalent α is given by

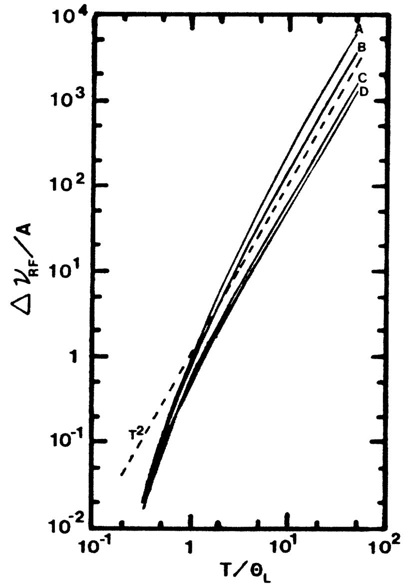

FIG. 5. Temperature dependence of the fracton contribution to the Raman broadening of spectral lines. The curves are numerical evaluations of the linewidth given by the second term in Eq. (5). In curve A: $\tilde{d}=\frac{4}{3}$, $\bar{d}=3$; curve B: $\tilde{d}=\frac{4}{3}$, $\bar{d}=2.5$; curve C: $\tilde{d}=1$, $\bar{d}=3$; curve D: $\tilde{d}=1$, $\bar{d}=2.5$. In each case we have taken $\Theta_F \gg T$. The quadratic dependence shown by the dashed line is included for reference.

$$\alpha \approx A\Theta_D^{10}/(\Theta_L^3\Theta_X^7) , \quad (6)$$

where Θ_X is the Debye temperature of this equivalent crystal. Using our observation that the homogeneous linewidth in the glass is about 10^{-4} $T^2\,\text{cm}^{-1}$, one obtains $\alpha \approx 10^2$ cm^{-1}, which is of the same order as observed for rare-earth transitions in crystals.[16]

Physically, the reason that the fractons produce a larger Raman contribution to the linewidth than found in crystals is that the density of states for the fractons at low frequency is substantially larger than would be found for Debye phonons. This can readily be seen from Eq. (3) since ω_L is comparable to ω_D, but $d=3$ while $\tilde{d} \approx 1$. The quadratic temperature dependence results from the non-Debye-type frequency dependence of the density of states and dispersion.

Implicit in the integration of the linewidth over the whole fracton spectrum is the assumption that the excited ions can in fact sample all of these excitations. With the localized nature of the fractons and the site selection of a FLN experiment, it is not obvious that this will be the case in all conceivable structures at all temperatures. For a glass whose structure is a single self-similar fractal, every site in the glass will sample many scales of length and essentially all scales of large l. The quadratic linewidth which we calculate is determined almost exclusively by fractons with $\hbar\omega < kT$ leading to the expectation that all the important fractons will be available at each site. Nevertheless, the various fractons may couple to the electronic states with different strengths. By using the *average* local strain in Eq. (2), the electronic matrix element $|\langle f \mid V_R \mid i \rangle|$ becomes an average over the mode couplings as well. This is also true of electron-phonon coupling in crystals in the equivalent approximation. The fact that our experiments show the same temperature dependence on both sides of the inhomogeneous profile supports the validity of this approximation for these glasses. However, since the magnitudes of the high- and low-frequency linewidths are different, this average must be taken with different weights in these two parts of the inhomogeneously broadened band. The gross difference between the LS and NS glasses suggests that the frequency dependence of this effective coupling constant is a structural property that might be predictable from the connectivity of the fractals in a more detailed model. For $T \ll \Theta_L$ the spatial distribution of fractons may become dominant and destroy the validity of this approximation.

Liu[17] has recently developed a model for the localized modes in which the glass structure is taken to have three-dimensional connectivity, but the strengths of the interatomic bonds are fractal, giving $\tilde{d}=1.5$ and $p=1$. The number of modes at low frequency is reduced relative to that obtained when the connectivity is fractal. Because of this, the homogeneous linewidth does not follow a simple power-law dependence in the temperature range of our experiments.

For Eu^{3+} with the resonant pumping conditions used in the experiments described here, the only possible direct phonon process which can contribute to the broadening of the spectral line is between the $^7F_0$ ground state and the components of the $^7F_1$ manifold which are split by 250 cm^{-1} in these glasses. The increased density of states for the fractons at low frequencies must come at the expense of their high-frequency density of states. Using the data reported by Selzer *et al.*[4] for YAlO$_3$:Eu^{3+}, we infer a direct-process contribution to the linewidth of $\approx 10^{-2}$ cm^{-1} at 100 K, leading to the conclusion that the direct process will be unimportant in these glasses at all temperatures studied here.

Since the submission of our original manuscript, Alexander, Entin-Wohlman, and Orbach[18,19] (AEO) have treated the problem of electron-fracton interactions. They assume the differences in "crystal field" to arise from the static fluctuations in density, on a scale large compared to atomic dimensions, that characterize the fractal geometry. Sites that are spectroscopically equivalent would also be geometrically equivalent in this sense. Since the fractons are localized vibrations, each site in the atomic network is exposed to only a subset of all the fractons; this subset being determined by the fractal mass distribution. There would then be a specific set of fractons that would dominate the two-fracton relaxation at each spectroscopically equivalent site. Considering the interaction with these dominant fractons, rather than averaging over all modes as we have done, they find a $T^{2.63}$ dependence for the homogeneous broadening. While the precise power law that governs our data can be varied by different choices of the residual inhomogeneous broadening, the power law proposed by AEO is too large to account for our results, except for the KG sample, where our data do not extend to high enough temperatures to minimize the effect of the residual inhomogeneous contribution. It seems to us that the restricted set of fractons allowed to interact with each site by the localization imposed by AEO may not be a good representation for all glasses. This is because glasses may have other contributions to the local crystal field than those imposed by fractal symmetry, and over the millisecond time scale of our experiments, dynamical modes besides fractons and Debye phonons (we think here of the ubiquitous TLS and optical modes) may alter the local fractal geometry. This being the case, an average over all fractons, as we have done here, may be a better approximation for glasses of the type studied here than demanding the full restrictions imposed by fracton localization. In other types of fractal systems the localization may be all important.

V. SUMMARY AND CONCLUSIONS

We have presented measurements on three structurally different glasses showing the homogeneous linewidth of the $^5D_0$-$^7F_0$ transition to have a quadratic temperature dependence between 10 and 300 K and the residual inhomogeneous broadening in the narrowed fluorescence to increase with increasing excitation frequency across the inhomogeneously broadened band. Using resonant pumping techniques, eliminated contributions to the data from overlapping Stark components and direct phonon processes. The results cannot be interpreted in terms of conventional processes involving electron-phonon interactions with Debye-type phonons. The observed behavior appears to be independent of host structure since it is qualitatively the same for the three glasses, whereas other details of the optical spectra are known to be quite different for these three materials, due to structural differences of the glasses.[10]

A simple model based on the fracton concept has been introduced to explain the behavior of the homogeneous linewidth. The increased linewidth and its quadratic temperature dependence are shown to be consequences of Raman broadening by the localized vibrational modes of the glass and their non-Debye-type dispersion and density of states. This model involves fractal behavior of the local structure, as opposed to bonding, and involves electron-phonon coupling modified by the fractal behavior as opposed to direct electron-TLS coupling. The tunneling modes of TLS do not appear to contribute to the line broadening at the high temperature range investigated here.

The experimental data and theoretical model presented here provide new information concerning the effects of local vibrations on the widths of spectral lines of impurity ions in amorphous host materials. Tests of the universality of the observed behavior of the temperature dependence of the homogeneous linewidth and the applicability of the model must await further experiments on different types of materials.

ACKNOWLEDGMENTS

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